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NANOINDENTATION AND TEM CHARACTERIZATION OF ION IRRADIATED 316L STAINLESS STEELS

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Abstract

Understanding the effects of extensive radiation damage in structural metals provides necessary insight for predicting the performance of those metals considered for application in the extreme radiation environment. Predicting mechanical performance after long term radiation exposure is of great importance to extending the life of current nuclear reactors and for developing future materials for the next generation of reactors. A combination of finite element modeling, nanoindentation, and TEM characterization were used to rapidly determine the microstructure and mechanical properties influences of ion irradiation on a standard 316L stainless steel sample. The results of this study found that ion irradiation and small scale mechanical property testing can be used to characterize extensive levels of radiation damage structure, only when significant consideration is given to ion irradiation depth, surface roughness, polishing condition, the irradiation temperature, and many other experimental parameters.

Introduction

For either the success of next generation nuclear reactors or the extension of current nuclear reactors significantly beyond their designed lifetimes, research is needed into the degradation of the microstructure and mechanical properties governing the performance of the reactor alloys exposed to extreme environments. Although the exact dose and temperature of these nuclear reactor plans may vary, the cladding and other structural components will be exposed to greater dose and at elevated temperatures. To accommodate these severe exposures, established cladding materials are being testing to longer exposure and new alloy compositions are being considered. [1-3] For neutron test facilities to reach 100 dpa level, exposure times often span several years, significantly limiting the down-select process of potential new materials for extreme radiation environments. Ion irradiation has been used as a technique to provide a look at the effect of irradiation damage on materials in a significantly shorter time frame. Previous work using proton irradiation provides a deep, broad, and relatively uniform defect profile compared to heavy ion irradiation; however, using this type of radiation still requires weeks to months to reach the 100 dpa level. [4] In this research, we investigated the feasibility of using heavy ion (nickel) irradiation and small scale mechanical testing to provide in a few days a first order screening technique for potential alloys that are being considered for advanced testing.

Finite Element Model

To determine if the effects of irradiation damage would be detectable by nanoindentation, an axisymmetric finite element model was created to simulate the nanoindentation experiment using a conical tip with an effective cone half-angle (19.7°) of a sharp 3-sided diamond Berkovich tip.

[5] The finite element mesh of the model is shown in figure 1A. The size of the substrate in this simulation is a 60 μm by 40 μm and contains a sub-surface hardened region, highlighted in red. The bottom of the substrate is fixed in the simulation and the hardened region is 2.5 μm thick situated between 2.5 and 5 μm beneath the substrate surface. It is included to capture the effect of a radiation hardened layer beneath the surface of a 300 series stainless steel sample. In the simulation, the diamond indenter has a Young's modulus of 1141 GPa and a Poisson's ratio of 0.07. The substrate has a Young's modulus of 200 GPa and a Poisson's ratio of 0.3. The coefficient of friction between the tip and the surface was set as 0.1. The yield strength of the substrate was set to 350 MPa. To provide a first order approximation of a radiation hardened region produced by ion irradiation, the region outlined in red was set to a yield strength of 700 MPa.

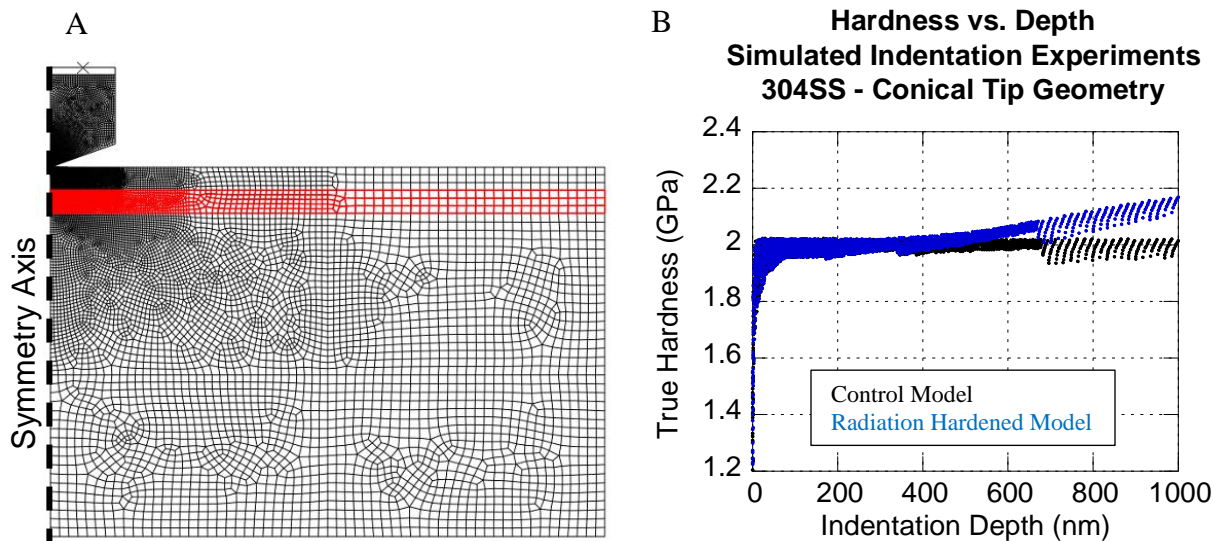


Figure 1. A) Finite Element Model of conical indentation of with an irradiation hardened microstructure. B) Simulated hardness for both control and irradiated conical nanoindents based on the FEM simulation

A simulated result demonstrating the effect of the subsurface hardened layer is given in figure 1B. It is a plot of hardness, i.e. load divided by projected contact area, vs. indentation depth. For comparison, the same result from simulation without a hardened region is also plotted in figure 1B. The comparison shows the increasing influence of the hardened region as the indentation depth increases, even though the maximum depth in the simulation is significantly less than the depth of the hardened region. This result suggests that the plastic zone ahead of the indentation tip begins interacting with the hardened region, at an indentation depth of about 500 nm. The hardness was found to increase as with increasing indentation depth suggesting that nanoindentation should provide an adequate technique to sample the radiation damage in the limited volume that is produced by ion irradiation.

Experimental Procedure

To investigate the feasibility of using high-energy heavy-ion irradiation that provides a first order simulation of neutron damage in advanced cladding materials, a variety of irradiation parameters and small scale mechanical testing conditions were investigated. All of the studies

discussed in this proceeding are based on nine 8 mm x 8 mm sample coupons that were cut and mechanically polished to a mirror finish from the same sheet of 316L AK Steel. The manufacture reports the tensile strength as 631 MPa the 0.2% offset yield strength as 331 MPa, and the Rockwell hardness as 82.[6] The final polish of the coupons was done using a vibratory polish using colloidal silica.

All of the samples were irradiated with 20 MeV Ni at 400 °C, 500 °C, and 600 °C at a dose rate of approximately 0.01 dpa/s by the 6MV Tandem accelerator at the ion beam laboratory at Sandia National Laboratories. Three total doses were performed at each temperature that were calculated by the Robinson equation [7, 8] to be equivalent to 10 dpa, 40 dpa, and 100 dpa. The damage distribution produced by these ion irradiation conditions were modeled using stopping and range of ions in matter (SRIM) program. [9]

Sets of 16 nanoindentation experiments were performed on each of the samples both in the irradiated region and in the control region significantly outside of the irradiated region. The experiments were performed using a three sided diamond Berkovich tip geometry operated in load control at a constant true strain rate to a depth of 1 μm . Deviation in the 16 nanoindentation per condition were used to provide the errors bars associated with each measurement [10, 11] TEM thin foils of the samples were prepared by traditional focused ion beam (FIB) lift-out technique followed by a low energy polish to limit the Ga implantation related defects. The TEM foils were observed using a JEOL 2100, a Phillips CM20, a Phillips CM30, and a FEI Tecnai under a variety of imaging conditions to best identify the defect structures created.

Results and Discussion

As predicted by the finite element model, a distinction was observed in the majority of the ion irradiated regions when compared to the set of baseline nanoindentation results, as can be seen in Figure 2. In the 316L coupon irradiated to approximately 10 dpa, the difference between the ion irradiated regions and the baseline was slight, falling within the error bars for the implants done at 500 °C and 600 °C. At approximately 40 dpa, a significant increase in hardness is observed between the baseline nanoindentation results and those of the 400 °C and 500 °C irradiation exposures. However, the hardness values for the material receiving a 40dpa exposure at 600 °C remained similar to the baseline. This trend is enhanced in the 100 dpa set of implants resulting in a 1 GPa difference between the hardness of material ion implanted at 400 °C and 500 °C compared with the baseline and the 600 °C implanted region.

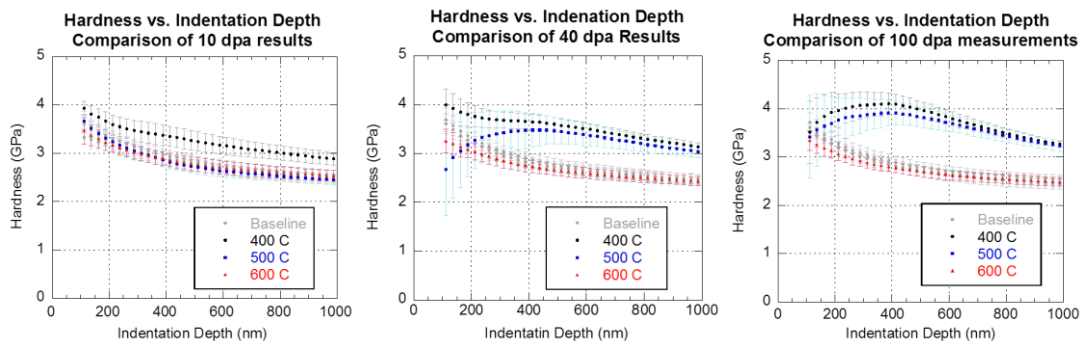


Figure 2. Hardness of the irradiated regions and control regions from nanoindentation as a function of temperature and dose

Initial analysis of the data raised a concern regarding the decreasing slope of the baseline hardness versus depth curves shown in Figure 2. To determine if this was a small scale effect or if the surface had been hardened during the metallographic sample preparation, the samples were electropolished. As seen in Figure 3, the electropolished baseline sample did not exhibit the same negative slope seen in the initial coupons suggesting that the negative slope seen was a result of work hardening that occurred in the relatively malleable 316L stainless steel. This work hardening appeared to be limited to the top 400 nm to 600 nm of the coupons and produced significantly less damage than the radiation damage produced in the majority of the coupons.

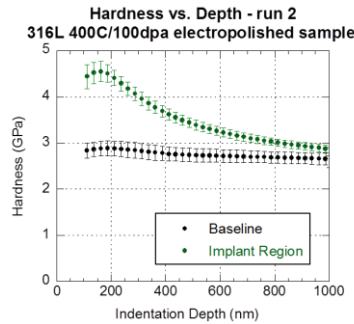


Figure 3. Hardness of the control and irradiated samples after electropolishing

To distinguish between radiation hardening from ion implantation and the work hardening from sample preparation, the data presented in Figure 1 is normalized to the baseline and is plotted again in Figure 4. The effect of increasing dose on the hardness ratio can be observed in both the 400 °C and 500 °C sets of coupons in the normalized data indicating that the effect of dose was significantly larger than the surface work hardening resulting from the vibratory polishing. Figure 4 also clearly demonstrates that there is minimal deviation between the baseline samples and the set of samples irradiated at 600 °C. As a consequence of the observation that surface and subsurface conditions can greatly affect nanoindentation results, it is strongly recommended that a baseline normalization and similar hardness ratio should be implemented in any technique to rapidly characterization of ion irradiation damage.

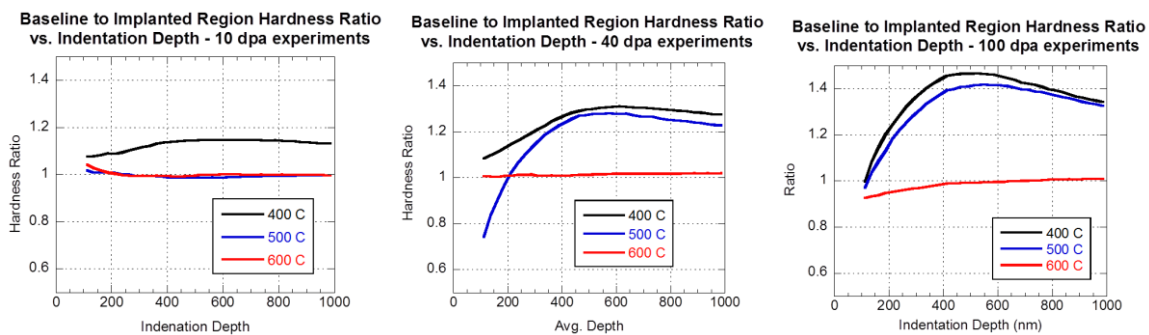


Figure 4. Hardness of the irradiated regions from nanoindentation normalized to the baseline hardness as a function of temperature and dose

An electron microscopy study of the ion irradiated regions was initiated to determine the microstructures that evolved from ion irradiation to approximately 10 dpa, 40 dpa, and 100 dpa at temperatures of 400 °C, 500 °C, and 600 °C. The FIB lift-out samples were cut beyond the ion implantation end-of-range permitting the comparison between the defect structures resulting from 1 keV Ga ion final milling and the 20 MeV Ni ion implantation. The defect density in the region only exposed to FIB damage was significantly less than that produced by ion

implantation, making it easy to distinguish the ion irradiated region in the films. Cross sectional TEM analysis indicated that the majority of the radiation damage in all of the films was present at the end of range of the ion beam, approximately 3.7 μm deep. The microstructure of ion implantation damaged regions included dislocation tangles, dislocation loops, small voids, and other small defect structures. The density and location of these defects were found to be a function of depth into the sample and ion irradiation conditions.

To compare the effects of various implantation dose and temperature, TEM bright-field micrographs of cross-sections were taken at a depth of 1.5 μm below the surface of the original coupon. This region was chosen as the damage profile predicted by SRIM to be relatively flat and it was significantly beyond the denuded region at the surface of the coupon. In comparing the micrographs, the expected trend of increased damage with increasing dose can be seen. The 10 dpa foils show isolated defect clusters that image as black clusters, whereas it is difficult in the 400 $^{\circ}\text{C}$ 100 dpa image to clearly identify any isolated defect structures. The formation of larger dislocation loops and voids in the high temperature implants provide an explanation for the observed nanoindentation results. It is well known that as defect density decreases the force need for a dislocations to pass through the defect field significantly decreases resulting in the decreased hardness between the coupons irradiated at 600 $^{\circ}\text{C}$ and those at 400 $^{\circ}\text{C}$.

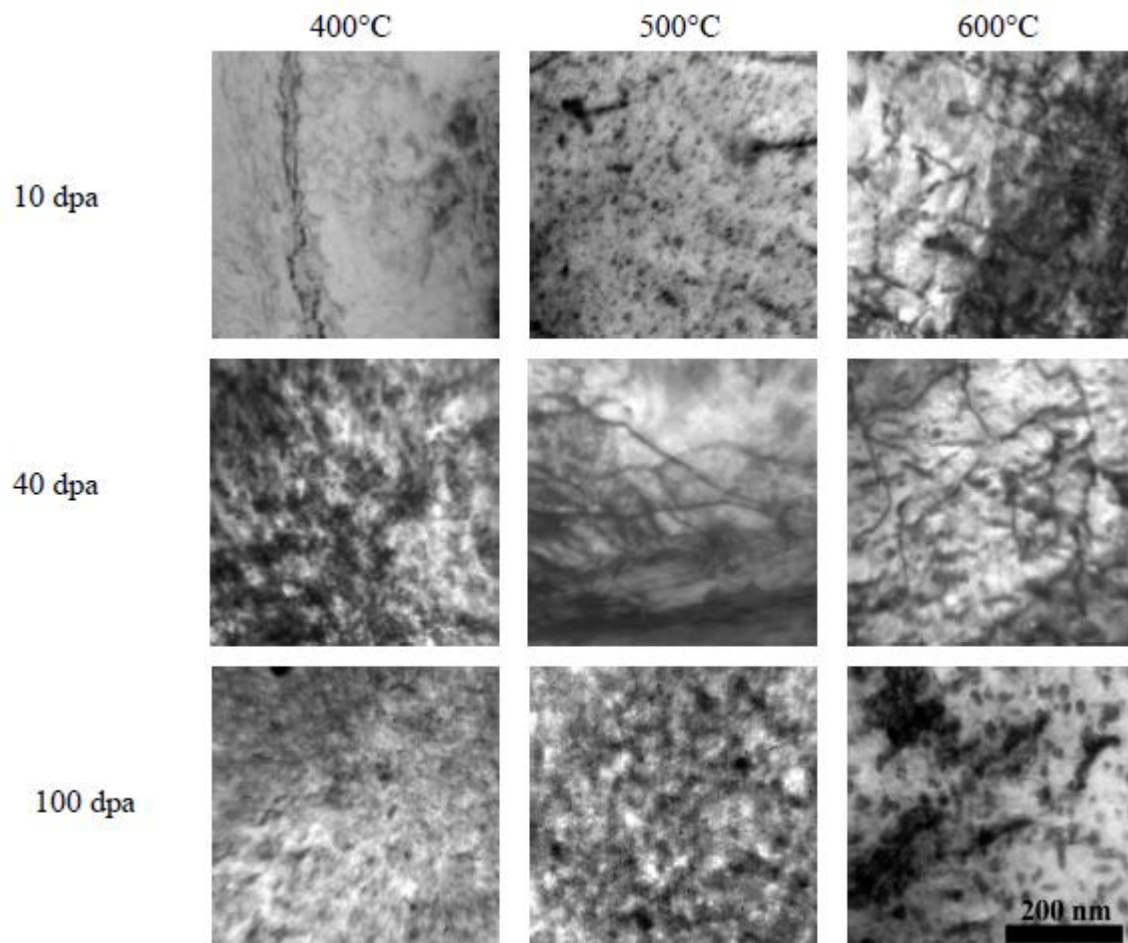


Figure 5. Bright field TEM images taken at 1.5 μm deep from the surface of the irradiated regions at the dose and temperature specified.

To further understand the effects of the ion irradiation on the 316L coupons, energy dispersive X-ray spectroscopy and mapping were performed on the TEM samples. For the majority of the samples, minimal heterogeneity was observed and in those cases was often associated with grain boundaries, as might be expected. However in the coupon irradiated to 100 dpa at 600 °C a significant amount of both Ni and Si solute segregation were observed to occur, as can be seen in Figure 6.

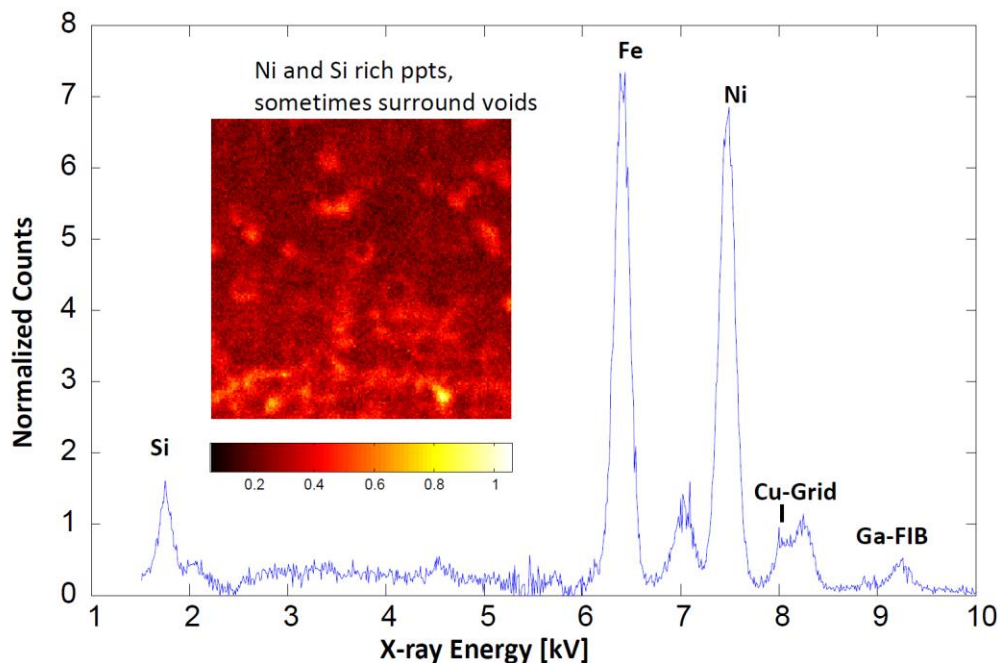


Figure 6. EDS spectra and map of the segregation in the 100 dpa 600 °C TEM sample. The map is 500 nm x 500 nm.

Although the post mortem TEM characterization provides insight into the microstructures that form from radiation damage in these samples, it provides little insight into the mechanisms by which this microstructure evolves under these conditions for such understanding in-situ observation of the microstructure is needed.

Although this technique shows promise as a very rapid technique to screen for materials that are resilient to extensive radiation damage, it raises many concerns that needed to be addressed if it is ever applied to advanced materials. The first concern is the volume of the small scale testing technique chosen. It must be significantly smaller than the irradiation volume, but significantly large enough to average over the heterogeneous microstructures of the sample. Nanoindentation was found to provide this capability in a rapid fashion and as such was chosen over other techniques like micropillar compression. The selection of the ion species and energy is important to provide heavy displacement damage, minimal compositional variation, and a deep flat damage profile. Although the choice of Ni did provide extensive displacement damage and minimal alteration in composition of the 316L steel, it did not provide an extremely deep and flat distribution of defects making it difficult to associate the recorded hardness with an observed microstructural region. In addition, the use of Ni ion beam has been indicated by others to have additional adverse effects when implanted into steels and thus provides a poor choice. [12] Finally, the use of heavy ion irradiation alone does not provide any insight into the bubble formation, swelling, blistering, and hydrogen embrittlement that are observed in neutron

exposure. A significant representation of any of these effects alone can result in the exclusion of a material system from consideration.

Current work is underway to better understand the microstructural evolution and the resulting effect on mechanical properties to further refine the proposed method to rapidly characterize to a first-order the viability of new structural metals for extreme radiation environments. A nearly in-situ scanning electron microscope (SEM) and electron back-scatter detector (EBSD) system on a Tandem accelerator end station has been established to characterize texture evolution as a function of dose. In addition, an in-situ ion irradiation TEM has been established at Sandia to permit fundamental studies of radiation damage evolution. This TEM and the coupon implanting chamber in front of it are being developed into a triple beam system for the simultaneous concurrent implantation of heavy ions, He, and D₂. Work is also underway to capitalize on the small scale nature of the irradiation and testing by reproducing this work on various 316L and HT9 based diffusion multiples. Finally, work to incorporate a distribution of hardness either from the distribution of various defect structures provided by TEM or through SRIM calculations into the FEM has been initiated.

Conclusions

The combination of ion irradiation, nanoindentation, and TEM analysis of FIB lift-out samples has been shown to be a rapid and efficient method to characterize a materials microstructural evolution and mechanical properties of 316L stainless steel with extensive radiation damage. Nanoindentation was found to be an adequate small scale mechanical testing technique that can rapidly characterize the effect of radiation damage in the limited volume produced by ion irradiation. In applying this combination of techniques care must be taken to account for the effect of sample preparation techniques for both the nanoindentation and TEM analysis. Despite the concerns that need to be addressed, the success of this study suggests that ion irradiation and small scale mechanical testing is a promising method for characterizing the microstructure and properties resulting from extensive radiation damage. This testing can provide a rapid screening technique of the various alloy composition and microstructures of minimal volumes that can enhance further and necessary studies using both proton beam irradiation and neutron exposure.

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References

1. Etienne, A., et al., *Atomic scale investigation of radiation-induced segregation in austenitic stainless steels*. Journal of Nuclear Materials, 2010. **406**(2): p. 244-250.
2. Odette, G.R. and D.T. Hoelzer, *Irradiation-tolerant Nanostructured Ferritic Alloys: Transforming Helium from a Liability to an Asset*. JOM, 2010. **62**(9): p. 84-92.

3. Sencer, B.H., et al., *Microstructural analysis of an HT9 fuel assembly duct irradiated in FFTF to 155 dpa at 443 degrees C*. Journal of Nuclear Materials, 2009. **393**(2): p. 235-241.
4. Was, G.S., *Fundamentals of Radiation Materials Science: Metals and Alloys*. 2007, New York: Springer-Verlag Berlin Heidelberg. 827.
5. Fischer-Cripps, A.C., *Review of analysis and interpretation of nanoindentation test data*. Surface & Coatings Technology, 2006. **200**(14-15): p. 4153-4165.
6. *AK Steel Corporation Metallurgical Test Report*, L.N. Brewer, Editor. 2008: Rockport, IN.
7. Oen, O.S., D.K. Holmes, and M.T. Robinson, *RANGES OF ENERGETIC ATOMS IN SOLIDS*. Journal of Applied Physics, 1963. **34**(2): p. 302-&.
8. Robinson, M.T. and O.S. Oen, *COMPUTER STUDIES OF SLOWING DOWN OF ENERGETIC ATOMS IN CRYSTALS*. Physical Review, 1963. **132**(6): p. 2385-&.
9. Ziegler, J.F., M.D. Ziegler, and J.P. Biersack, *SRIM - The stopping and range of ions in matter (2010)*. Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms, 2010. **268**(11-12): p. 1818-1823.
10. Kraft, O. and C.A. Volkert, *Mechanical testing of thin films and small structures*. Advanced Engineering Materials, 2001. **3**(3): p. 99-110.
11. Oliver, W.C. and G.M. Pharr, *Measurement of hardness and elastic modulus by instrumented indentation: Advances in understanding and refinements to methodology*. Journal of Materials Research, 2004. **19**(1): p. 3-20.
12. Edwards, D.J., et al., *Nano-cavities observed in a 316SS PWR flux thimble tube irradiated to 33 and 70 dpa*. Journal of Nuclear Materials, 2009. **384**(3): p. 249-255.